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Hydrodynamic Scaling in the Randomly-Networked Liquids of Silica and Silicates

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熔融ケイ酸およびケイ酸塩における流体力学的緩和時間は非常に長く、分子動力学 (MD) シミュレーションで計算可能な範囲を越えている。従って、これらの液体の粘性係数を MD シミュレーションから算出すると、有限周波数における値が求まるのみである。本研究では、有限周波数から零周波数への粘性係数の移行を、流体力学的スケーリングにより論ずる。新たに導入される流体力学的指数の組成、圧力による変化から、粘性係数の変化が推定され、その結果は実験事実と整合的である。この指数は、熔融ケイ酸およびケイ酸塩中のランダム・ネットワーク構造により決定される。

Molecular dynamics (MD) simulations innovate in probing the collective dynamics of the constituent atoms which determines hydrodynamics in liquids. The shear viscosity is the transport coefficient of the transverse current in the hydrodynamic limit, *i.e.* in infinite wavelength (or, zero wavenumber) and infinite time (or, zero frequency) relative to an atomic scale, and can be computed by tracing the correlation of the transverse-current modes over the shear relaxation time¹⁾. This computation is, however, hardly tractable in the cases of inorganic glass-forming liquids of silica and silicates, because of the long shear relaxation time, typically tens nanoseconds or longer. In these cases, merely the extension of the shear viscosity to finite frequency can be computed²⁾. The finite-frequency shear viscosity is smaller than the zero-frequency shear viscosity.

Molten silica (SiO_2) and silicates (e.g. Mg_2SiO_4) have the randomly-networked structure which is connected by the silicon-oxygen bonding and is, on the other hand, terminated internally by 1- or 2-valent cations. The slow shear relaxation is induced by the long-lived modes which are generated by the strong non-linear coupling of modes in the randomly-networked structure. The small magnitude of the 'bare' shear viscosity at finite frequency is, therefore, 'renormalized' to the large magnitude of the 'dressed' shear viscosity at zero frequency due to the strong mode-coupling in the random network.

The renormalization of the shear viscosity from finite frequency to zero frequency is realized by the hierarchical coarse-graining³⁾. The bare shear viscosity, or the transverse-current memory function $K_T(k, \omega)$ at wavenumber k and frequency ω , is coarse-grained by integrating its convolution to the Lorentzian window of the finite half-width Γ ;

$$K_T(k, \omega; \Gamma) \equiv \int_{-\infty}^{\infty} \frac{d\omega'}{\pi} \frac{\Gamma}{(\omega - \omega')^2 + \Gamma^2} K_T(k, \omega'). \quad (1)$$

The coarse-graining hierarchy is then obtained from Eqn (1) with decreasing Γ . $K_T(k, \omega; \Gamma)$ with small Γ exhibits the discernible peak at $\omega = 0$, and this central peak grows steeply with decreasing Γ . The steep growth of the central peak indicates the intensification from the bare shear viscosity to the dressed shear viscosity due to the renormalization correction at $\omega = 0$. It can be described by a power law;

$$K_T(k, \omega = 0; \Gamma) \sim \Gamma^{-w_T}. \quad (2)$$

Eqn (2) defines a novel exponent, w_T , which is a 'hydrodynamic' exponent, characterizing the approach from the bare shear viscosity to the dressed shear viscosity. A larger w_T indicates the larger renormalization correction, and therefore does the larger magnitude of the dressed shear viscosity.

The hydrodynamic exponents w_T are obtained from MD simulations on molten SiO_2 (silica) and molten Mg_2SiO_4 (forsterite, or magnesium-olivine) at various pressures. The results show that w_T changes differently by composition, and indicate the cross-over pressure, $P_c \sim 2.5$ GPa, and the critical value of a hydrodynamic exponent, $w_T^c \approx 0.6$. In molten SiO_2 , on the one hand, w_T decreases above P_c from $w_T > w_T^c$ to $w_T < w_T^c$. In molten Mg_2SiO_4 , on the other hand, w_T increases above P_c from $w_T < w_T^c$ to $w_T > w_T^c$. w_T then converges to w_T^c at ~ 10 GPa in both liquids.

The pressure-induced decrease of w_T in molten SiO_2 and increase in molten Mg_2SiO_4 indicate the negative and the positive pressure-dependences of the dressed shear viscosities, respectively, in consistency with the empirical relation of the pressure-dependence of the shear viscosity to composition, or to the network connectivity⁴⁾. The converged hydrodynamic exponent w_T^c suggests its mean-field-like behaviors. The cross-over pressure $P_c \sim 2.5$ GPa coincides with those of the minima of the sound speed and the elastic moduli of vitreous silica⁵⁾, and suggests that the pressure-induced change in the randomly-networked structure, especially the densification of the 'packing' of atoms, is responsible for the enhancement or hinderence of the mode propagation, and is accordingly for the cross-over of the shear viscosity.

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